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G. I. Stegeman

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This final report presents research into the effects of intensity-dependent refractive indices in optical fibers and thin films. Discussed are photosensitive gratings; second-harmonic generation in fibers and at surfaces; the theoretical investigation of all-optical switching; all-optical switching in fibers; and two-photon absorption and color center dynamics.

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EXECUTIVE SUMMARY

This report addresses the three-year program of research on nonlinear effects in fibers and, to a much smaller extent, on second-order nonlinear optics at surfaces.

Nonlinear Effects in Fibers

The physics and applications of second-harmonic generation (SHG) and photosensitive gratings in fibers were investigated, both theoretically and experimentally. All-optical switching was demonstrated in rocking filter and grating fibers, and various switching schemes and their limitations were analyzed. The importance of two-photon absorption and color centers as limiting factors in all-optical switching was investigated.

After a standing wave was initially established through reflection at the ends of photosensitive GeO-doped fibers, highly efficient distributed feedback gratings were written in the fibers by blue-green light. By measuring the grating strength's dependence on wavelength, the concentration of Ge E' centers created, and the absorption, the origin of the index change was established as two-photon absorption into the UV bleachable absorption band of an oxygen-deficient germania site in the glass matrix.[1,2] Careful measurement of grating growth with time revealed slow oscillations that eventually resulted in saturation of the grating reflectivity.[3] This time evolution was successfully modeled by a bleaching of the two-photon absorption band.[3,4] We believe that we understand, and can now model, the writing of photosensitive gratings in such fibers.

Two applications of these gratings were investigated. The group velocity dispersion associated with the periodic structure, as measured with a Mach-Zehnder interferometer, produced regions in the visible where a negative group velocity dispersion is possible for fiber modes.[5] In principle, it should be possible to propagate solitons in the visible and achieve pulse compression. It was also shown that the writing process's high sensitivity to vibrations can be applied as a vibration sensor, with a sensitivity of a few tens of angstroms.[6]

Anomalous SHG in glass optical telecommunication fibers occurs when a nominally centrosymmetric glass fiber "self organizes" into an efficient, phase-matched second-harmonic generating structure after exposure to a YAG laser beam. A number of experiments were performed to elucidate the origin of this self organization. Detailed polarization studies of the process, infrared erasure of the SHG phase-matching gratings, and the temperature dependence of the writing process all point towards a photorefractive interpretation for the process.[7-9] The origin of the large DC electric fields required for the effect remains unexplained.[10,11] Results of experiments exploring the SHG signal's dependence on length, and its variation with green seed power, also remain unexplained.[12]

All-optical switching, that is, the use of high intensities to change the throughput response of a fiber device, has been demonstrated in two systems.[16-19] A rocking filter fiber normally rotates the plane of polarization 90° by matching a periodic strain (produced by twisting) to the birefringence. At high intensities, the synchronism is lost and the output polarization is the same as the input polarization. Picosecond pulses have been switched.[16-18] High intensities have also been used to break the Bragg condition for a fiber grating, switching the reflection coefficient.[19]

Extensive theoretical work has been performed to gain an understanding of the limitations on all-optical switching, to discover how these limitations might be circumvented, and to explore new problems introduced in the investigation.[20-31] The price paid for a sharp switch is that it will operate near some spatial or temporal instability.[20-22] Competing effects, such as parametric amplification and modulation instabilities, can also occur.[21,22] Limits on switching are also brought by pulse breakup, and dispersion limits the minimum pulse lengths that can be used.[23] One way to overcome these limitations may be to use temporal solitons, which would circumvent pulse breakup and pulse spreading.[24] The use of cross-phase modulation to stabilize soliton-like pulses in the visible region has been proposed.[25] Problems with soliton inputs, such as polarization instabilities, exist, however.[26-29] For the rocking filter fiber in particular, wavelength-selective switching of soliton inputs is possible.

Two-photon absorption and its effects on all-optical switching devices were investigated. A general material figure of merit was deduced and tested for lead-doped glasses with high nonlinearities.[32-35] Two-photon absorption posed a serious problem in the visible. In addition, color-center formation produced significant effects, similar to those obtained from a true two-photon response, but much slower.

Other Areas Investigated

Surface second-harmonic generation was used to study the microstructure of films fabricated by vacuum deposition and of Langmuir-Blodgett (LB) films.[12-14] The crystal symmetry and net orientation of monolayer crystallites deposited by the LB technique were measured, as was the orientation of columns in films deposited in vacuum.

To better understand the potential of all-optical switching, experiments were undertaken and calculations made for semiconductor materials. The switching figure of merit was calculated from band-filling nonlinearities for various semiconductors. GaAs and CdTe were optimum materials. To prepare for future work on all-optical switching in active fibers, a new technique for measuring the linewidth enhancement factor in an amplifier was developed. The technique was applied to a semiconductor in this first test experiment.

RESEARCH ACHIEVEMENTS

Photosensitive Gratings

The writing of gratings into GeO-doped glass fibers by an argon-ion laser was reported more than a decade ago. The understanding of this effect lies in the assumption that the initial intensity standing wave pattern in the fiber (arising from the weak Fresnel reflection from the output end of the fiber) is somehow translated into a permanent, periodic modification of the index of refraction of the glass. Such a periodic structure would act as a Bragg-matched phase grating, causing additional reflection of the laser beam until some steady state is reached, with much of the laser beam reflected from the fiber. Two major questions arise: 1) how does a low-loss communications fiber absorb significant amounts of laser light to make such a transformation possible, and 2) what is the mechanism by which the index of refraction is permanently modified?

The steady-state modulation depth of the index grating is a quadratic function of the laser power during the preparation stage, suggesting a two-photon absorption mechanism. There is a broad defect absorption band at 245 nm, which would be optimally situated for two-photon absorption of the laser. This band has been shown to arise from oxygen vacancy defects of GeO_2 present in the core of the fiber. We have shown by ESR that the absorption of light into this band results in the creation of Ge E' centers. A clear correlation among the wavelength dependence of the number of E' centers created, the grating amplitude (quantified by the reflectivity) and the absorption spectrum of the oxygen vacancy defect accessed by two-photon absorption has been observed.

The mechanism through which the index is permanently modified has been the subject of intense debate over the last decade. We have demonstrated that bleaching of the absorption band is responsible for the index change, by monitoring changes in the average index of the fiber during the growth process with a single-mode He-Ne probe laser. The fiber in effect acts as a Fabry Perot interferometer, which is very sensitive to average index changes in the fiber. We have observed that the probe transmission is modified through what we believe are changes in the index of the glass, consistent with a bleaching of the 245-nm band.

Such a model is manifestly local in nature. Expressed in the literature are doubts that a local model could ever lead to sustained growth of a grating, rising from the phase of the grating relative to the intensity standing wave pattern. We have demonstrated through computer simulation that such growth is, in fact, possible, and have identified the falacy in the argument. Furthermore, we predicted and have observed one consequence of the bleaching model: slow oscillations of the transmission of the Argon laser beam during the growth process. These oscillations arise from a Fabry Perot effect as the laser modifies the average index of the glass.

While studying these photosensitive gratings we observed what at first appeared to be noise on the growth curves. Similar noise had been observed previously. We identified the origin of these oscillations as mechanical vibrations coupled to the fiber. Because the photosensitive gratings are long and weak, they are spectrally very narrow (with a bandwidth ~10E-6), hence the transmission of the laser beam is highly sensitive to variations in length as small as 50 nm. We have, therefore, proposed and demonstrated that such grating structures could be useful as highly sensitive passive all-optical vibration sensors.

We measured the wavelength dependence of the amplitude and phase response of a photosensitive grating by introducing the written fiber into one arm of a Mach-Zehnder interferometer. The other arm holds a virgin fiber as reference. From these measurements we were able to determine the maximum change in the refractive index. We were also able to show, from the phase response, regions of negative group velocity dispersion of the type that produce pulse compression in the visible. Computer modeling has established the limits on pulse compression that is feasible using such gratings, as well as the existence of solitons in those gratings.

Second-Harmonic Generation in Fibers

Second-harmonic generation (SHG) in some glass optical fibers occurs when they are illuminated by intense 1060-nm pulses. The writing process can be speeded up substantially by adding a weak green seed pulse. Although numerous models explaining this effect are now in the literature, all are far from satisfactory. There are also contradictions in experimental results, which we believe are inherent to the use of multimode fibers. Our experiments were carried out with single-mode fibers.

One of the models proposed for second-harmonic generation is a photorefractive effect. In this model charges created by two-photon absorption of the second harmonic light into the 245-nm band move in response to an optically created DC field. The first question posed was whether this field arose strictly from a mixing of the harmonic field with the fundamental field through a third-order nonlinearity. The magnitude predicted from this model would be a few volts per centimeter. Our first experiment was aimed at modifying the fiber preparation process, by applying a strong DC field to interfere with the field postulated to be created inside the fiber. No effect was observed up to a field strength of 10^4 volts/meter. This result showed that the internal fields used in moving charges are much larger than the initial model predicted.

Other experiments were performed specifically to test the photorefractive model. For example, if the formation of $\chi^{(2)}$ is essentially an EFISH effect, that is, a strong DC field acting in conjunction with the third-order nonlinearity $\chi^{(3)}$, then there are specific predictions for the ratios of the induced coefficents for the $\chi^{(2)}$ tensor. These ratios were measured by choosing appropriate combinations of the input and output optical field polarizations. Results were found to be in perfect agreement with the EFISH predictions. While this does not rule out other models, it would be completely fortuitous for another model to give the same predictions.

Within the framework of a photorefractive model, it should be possible to bleach out the phase-matching grating that gives rise to efficient SHG. Bleaching can occur by absorption of green light through two-photon absorption into the 245-nm range, releasing the trapped charges which form the phase-matching grating. This model predicts a fourth-power dependence on the infrared power for the bleaching, just as observed experimentally, yielding additional evidence for the photorefractive model.

Another key question addressed is the temperature dependence of the effect. Wide classes of models predict a strong temperature dependence of the preparation stage. Models in which the orientation of defects give a macroscopic $\chi^{(2)}$ would predict that the nonlinear grating would be enhanced if written at low temperatures, whereas photorefractive models would predict the opposite. It was reported recently in the literature that preparation of the fibers at liquid-nitrogen temperatures has no effect on the final SHG conversion efficiency, apparently a confirmation of a light-induced delocalization. To the contrary, our results show a strong temperature dependence, exactly that predicted on the basis of a photorefractive model.

We also investigated the spatial rate of growth of the SHG signal as a function of distance from the input end of the fiber, under a variety of conditions for the seeding beam. A great deal of data on the seed-power dependence of the SHG saturation was obtained, as was data on the position from the input end of the fiber, and on other parameters. A model for the dynamics of the grating growth is needed.

All-Optical Switching in Fibers

All-optical switching offers the possibilities of performing logic and of routing information on a sub-picosecond time scale. The output state of an all-optical device depends essentially on the input power level. This is only possible if the response time of the nonlinearity is very fast on the time scale of the desired pulses. One option is to use glass in fiber form, thereby compensating for the weak nonlinearity with the long propagation distances available. We have investigated all-optical switching in a number of fiber geometries.

Experiments on all-optical polarization switching in birefringent rocking-filter fibers were performed. These fibers were made by Roger Stolen at ATT Bell in Holmdel. The birefringent fibers are periodically twisted in the pulling process, with a periodicity equal to the birefringence beat length at a particular wavelength in the red. The effect of this twisting is to rotate the plane of polarization in the fiber with propagation distance. The fiber is cut to produce a 90° rotation at low input powers. At high powers, the index change caused by the intensity-dependent refractive index mismatches the birefringence beat length from the twist period, reducing the net rotation of the plane of polarization. At some power the net rotation produced becomes small, and the output light appears in the same state of polarization as the input light, promoting all-optical switching.

All-optical switching was demonstrated in the rocking-filter fibers with picosecond pulses. The process was modeled through coupled-mode theory, and good agreement with experiment was obtained.

We also investigated all-optical switching in distributed feedback gratings in optical fibers. The gratings were written with an argon-ion laser operating at 515 nm. The reflectivity for a weak 515-nm probe beam was approximately 50%. When a high-power pulse at 1060 nm was coupled into the fiber, transmissivity rose to approximately 100% over the duration of the picosecond pulse. The explanation of this observation is very simple. A high-power pulse modifies the refractive index and therefore disrupts the usual Bragg condition for the grating. The grating essentially "dissappears" over the duration of the pulse.

Theoretical Investigation of All-Optical Switching

Various calculations were performed in the area of fiber nonlinear optics. We limit the discussion to those calculations that affect all-optical switching directly.

The problem of polarization switching in a rocking-filter fiber was addressed, to determine the limitations of this device. In our experiments we noted an unexpected broadening of the switched pulses at high powers. We analyzed the propagation of short pulses in fibers using a Beam Propagation calculation that includes the effects of self- and cross-phase modulation. For the rocking-filter, the cross-phase modulation term is comparable to the self-phase modulation terms, and cannot be neglected. In the normal dispersion regime where our experiments were performed, we found that pulse spreading from phase modulation places a limit on the shortest pulse that can be switched efficiently. For typical fibers one meter long in the visible, as used in previous switching experiments, this limit is a fraction of a picosecond. This result explains the observed broadening. It also indicates that phase modulation can be a limitation in such fiber devices. Phase modulation can be reduced if shorter fibers are used, but these require higher switching powers, an unfortunate tradeoff.

A significant limitation on all-optical switching is pulse breakup. A pulse contains a continuous distribution of powers, ranging from zero to the peak power. As a result, different portions of a pulse will switch with different efficiencies, resulting in pulse breakup and incomplete switching. We postulated a solution to this problem for a nonlinear directional coupler, and verified it numerically in the anomolous dispersion regime. Temporal solitons are known to propagate in fibers without pulse spreading under appropriate conditions. Furthermore, the nonlinear phase shift is the same over the full pulse envelope. The switching of soliton input pulses in a nonlinear directional coupler was simulated using Beam Propagation codes. We found that soliton inputs, with the coupler coupling length adjusted to be equal to the soliton period, led to essentially complete switching of the pulse. Pulse breakup no longer occurs and the pulse switches completely, just as if it were a particle. A stability analysis established the range of inputs that yield stable outputs.

These calculations were extended to the case of the rocking filter fiber by including the effects of cross-phase modulation in birefringent fibers. In this case, soliton switching is more complicated. In fact, oscillations in the switching characteristics are predicted, because instabilities and branching of the orthogonal normal modes occur. Nevertheless, to a first approximation, soliton switching occurs, though not as cleanly as for non-birefringent fibers. A rocking filter fiber was also considered a candidate for wavelength-selective routing of optical solitons. In particular, we performed numerical calculations in which the input energy is fixed but the wavelength varied. Results show a large range of phenomena, but of most interest was that we did produce wavelength-selective routing of optical solitons. This finding suggests a new possibilty for all-optical signal processing based on wavelength-selective switching.

Cross-phase modulation between two pulses present simultaneously in a fiber produced novel results. Because the cross-phase modulation term is twice the usual phase modulation term, the phase modulation produced by the second beam is always larger than that of the self-phase modulation of the beam itself. This phenomenon allows a soliton to propagate in the normal dispersion regime, provided that it is accompanied by a dark soliton in the anomolous dispersion regime. This finding could have some interesting ramifications. Further work established limiting factors, such as grey soliton formation and polarization instabilities.

Two-Photon Absorption and Color Center Dynamics

We addressed the manner in which two-photon absorption affects an all-optical switching device. The imaginary part of $\chi^{(3)}$ gives rise to two-photon absorption which, in general, is undesirable, as it in turn leads to a significant loss mechanism. We derived a simple, dimensionless criterion which, for a given material, depends only on wavelength. The criterion tests a material's suitability for all-optical switching, by determining whether complete switching can be obtained without significant loss.

We investigated a lead glass fiber (high lead oxide content in the core and cladding) obtained from Bellcore as a potential medium for all-optical switching. Lead bulk glasses exhibit a high nonlinearity and have a known two-photon absorption coefficient at visible wavelengths. Our criterion was badly violated throughout the visible. On the other hand, we were able to demonstrate that the fiber makes an excellent optical limiter in the visible, requiring only on the order of 10 Watts peak for substantial limiting action. This limiting is a direct consequence of the strong two-photon absorption. Our criterion has found applicability in materials as diverse as organics and semiconductors, in which the role of two-photon absorption has become a key issue.

While studying two-photon absorption in the lead glass fiber, we discovered a large effective two-photon absorption attributable to color centers. We successfully modelled the interaction of the laser light with the lead glass, assuming that color centers are created through two-photon absorption of the laser, followed by a bleaching of the color centers through single-photon absorption of the laser. To account completely for the data, we postulated two species of color centers. The results were in good agreement with results of electron spin resonance studies of lead glasses reported in the literature.

When a steady-state distribution of color centers is achieved, the color centers can be shown to behave as if the glass has an even larger two-photon absorption coefficient than would otherwise be apparent. This finding limits the usefulness of the material for all-optical switching, just as true two-photon absorption does. While it is generally true that the color-center dynamics respond slowly (as opposed to true two-photon absorption, which responds instantaneously on the time scale of the laser pulse), these effects are cumulative. With prolonged use, color centers can render a material as useless as true two-photon absorption can. (Note that two-photon absorption in semiconductors leads to the generation of real carriers and hence can lead to effects on the time scale of carrier decay.)

Second-harmonic Generation at Surfaces

We have worked to develop second-harmonic generation (SHG) as a non-invasive probe of thin-film and Langmuir Blodgett (LB) monolayer microstructures. In SHG, a laser beam of frequency ω is incident upon the film to be probed, and the second-harmonic light at 2ω is detected by a photomultiplier tube. We use this technique because second-harmonic generation is relatively lacking in the supporting substrate, which is usually isotropic, yielding a surface sensitivity that is ideal for studying thin films. Furthermore, because SHG is mediated through tensors of the third (and sometimes fourth) rank, far more information about the symmetry of a film exists than does information about linear optics.

We demonstrated that SHG could be sensitive to the columnar growth of the microstructure of an inorganic thin film. Deposition of 0.5-μm-thick Al_2O_3 films on tilted glass substrates results in tilted column morphology. This microstructure has a global C_{1v} symmetry, which yields a highly characteristic signature in SHG.

To further investigate the sensitivity of SHG to columnar structure, we investigated ZnS films. ZnS films deposited by thermal evaporation are polycrystalline, with a superimposed columnar microstructure. If the orientation of the crystallites within the columns were random, the SHG signal would arise from the columns alone, and allow confirmation of theoretical predictions of the magnitude of the signal versus the film deposition angle. In our early attempts we found that preferential orientation of crystallites within the columns tended to dominate the signal, thereby masking the desired effect.

We also investigated monolayer LB organic films deposited on glass substrates. The films were prepared in-house, and at the Max Planck Institute for Polymeric Research in Mainz, West Germany, by Professor Wolfgang Knoll, through a successful collaboration. Prior to our work it was generally believed that crystallites are randomly distributed with respect to position and orientation on the water surface of the LB trough, and that these characteristics are transferred to the substrate during the dipping process. Our work was substantially facilitated by the fortuitous discovery (through SHG) that the J-aggregates, ellipsoidal crystallites on the order of 100 μm in diameter, are not randomly oriented on the water surface of the LB tank. Instead, the J-aggregates are slightly ordered during the isothermal compression phase, which preceeds their transfer to the glass substrate. This discovery itself has significance for a wide range of LB films, whenever the microstructure of the film can affect performance in device applications.

We measured the SHG signal on reflection from the monolayer. The incident beam was 45° from the surface normal, and the sample was rotated about the surface normal. The SHG angular symmetry obtained is indicative of C_2 crystal symmetry among the crystallites, confirming for the first time the "brickstone" model. This measurement was possible because the SHG process and the C_2 crystal groups both exhibit an even symmetry. As a result, even if the crystallites are randomly orientated, a net angular variation in the SHG persists.

Other Research

It is desirable that switching operations be cascable, that is, that the output of one logic gate or switching operation can be the input to the next. This scheme requires either lossless switches (fiber switches are as close as one can get to lossless switches) or switches with, or followed by, gain. Within that context we have attempted to gain a better understanding of the tradeoffs between index change and gain in an optical amplifier. The ultimate goal is to investigate Er-doped fiber amplifiers obtained from NTT.

To begin, we developed a Mach-Zehnder interferometer for measuring phase shifts in an electrically pumped InGaAsP amplifier. The ratio of index change to gain, the linewidth enhancement factor, is a critical parameter of amplifier performance. Before, this parameter had always been inferred from indirect measurements. We were able to measure the wavelength dependence of this parameter directly around 1.55 μm. Measurements for erbium-doped fibers are next.

PERSONNEL

George Stegeman, Faculty
Victor Mizrahi, Faculty
Ulf Osterberg, Faculty
Ewan Wright, Faculty
Bill Banyai (student)
Chris Krautschik (student)
Chai-Pei Kuo (student)
Brian Batdorf (student)
Ken DeLong (student)
Sophie LaRochelle (student)
Kelly Simmons (student)
Stephan Puderbach (visiting student)
Dave Heatley (student)
Stephan Trillo (visiting from Ugo Bordoni in Italy)
Stephan Wabnitz (visiting from Ugo Bordoni in Italy)
Dr. Yoshinori Hibino (visiting from NTT in Japan)
Professor Wolfgang Knoll (collaborator at Max Planck in Mainz)
Professor John Sipe (collaborator at the University of Toronto)

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